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(54) Title: COMPOUNDS FOR MANUFACTURING CROSS-LINKED PRODUCTS WITH A MATT SURFACE APPEARANCE, GREAT SOFTNESS AND HIGH THERMAL DIMENSIONAL STABILITY

(57) Abstract: The invention realizes a compound for manufacturing cross-linked products with great softness and thermal stability made by injection moulding, consisting of a mixture comprising: master polymers such as ethylene vinyl acetates (EVA), or polyethylene (PE) and their mixtures, otherwise rubber, ethylene-propylene (EPR); cross-linking agents, such as peroxides or similar; foaming agents, such as azodicarbonamides or similar. In addition the compound presents butadiene-acrylonitrile copolymers, even partially cross-linked.

COMPOUNDS FOR MANUFACTURING CROSS-LINKED PRODUCTS WITH A MATT SURFACE APPEARANCE, GREAT SOFTNESS AND HIGH THERMAL DIMENSIONAL STABILITY.

5 The invention concerns a compound for manufacturing foamed cross-linked products especially suited for producing items with a matt surface appearance, very soft and thermal dimensional stability.

Cross-linking polyethylenes (PE) and ethylene vinyl acetates (EVA) through the use of peroxides is a known technology for manufacturing injection moulded products such as, for example engineering articles, footwear, wheels,
10 etc.

The use of cross-linked products allows to obtain, against thermoplastic materials, polymers with greater physical and mechanical properties together with high heat resistance.

Cross-linked products can be basically divided into two types, compact and
15 foamed, which can reach very low minimum densities of around 100 Kg/m³.

It is mainly in the foamed version that these products demonstrate the positive contribution of cross-linking in their mechanical properties and thermal resistance.

It is these properties that have made cross-linked, foamed EVA products so
20 successful in the manufacture of soft products, while cross-linked PE products have proven their success in the manufacture of more rigid products.

The manufacturing process of any of the product can basically be divided into two stages.

In STAGE ONE the mixture or so-called compound is prepared with the
25 components required to obtain, in the final transformation process, a product that complies with the desired mechanical properties.

The compositions of known compounds generally comprise:

- Master polymers such as EVA and PE mixtures, or Ethylene-propylene rubbers (EPR), otherwise low and high density polyethylenes, having
30 various Melt Flow Index (MFI) levels, or ethylene vinyl acetates with different MFI's and different vinyl acetate contents;
- Peroxides, which are cross-linking agents that initiate activity at a given temperature, such as dicumyl peroxide, Bis (tert-butylperoxypropyl) benzene, 2,5 - Bis (tert-butylperoxy)- 2,5- dimethylhexane;
- 35 - Coagents, which block the secondary reactions of peroxides optimising

cross-linking, such as triallylcyanurates, triallylisocyanurates, ethylene glycol dimetacrilate and trimethylolpropane trimetacrilate;

- Foaming agents such as various forms of azodicarbonammides, which generate gas through their thermal destruction such as the types known industrially as Porofo® and Genitron®;
- Lubricant agents, which assist ejection of products from the mould and limit friction in the injection machinery, such as stearin and stearates;
- Anti-degradation agents, which protect the polymer from degrading during the manufacturing process, such as 6 – tert –butyl-m-cresol;
- Kickers, which control the degradation temperature of the foaming agents, such as metal oxides for example Zinc or glycols;
- Various forms of filler, such as calcium carbonates or silicas, are implemented both as nucleating agents to favour uniform dispersion of the components and gases, and to reduce costs in the mixture;
- Pigments, which are used to die the mixture;
- Oils that plastify the compound and/or reduce volatile dust in the mixture being processed.

One known method for manufacturing the compound prescribes that, after these ingredients have been adequately batched, they are mixed in special mixers until a uniform mixture is obtained, which is then fed into extruders that melt and blend the components in order to obtain a mixture that has all the additives finely dispersed in the master polymer/s.

Special care is taken with the extruders, which have to be kept at a temperature that does not initiate peroxide activity and does not decompose the foaming agents.

So at this stage no reaction is initiated, which makes this cycle exactly the same as the manufacturing and transformation process for thermoplastic composites.

The resultant final product is to all effects a thermoplastic ready to cross-link and foam if taken to the right temperature.

Another known method of manufacturing differs from the former in that the preliminary stage does not prescribe the preparation of a uniform blend.

This method uses an extruder equipped with weight batchers that first supply the extruder with polymer that is melted and then the various additives, which are amalgamated in the mass of molten polymer thereby creating the uniform

compound described previously.

Another method of manufacturing the compound, usually used in rubber manufacturing, prescribes the hot preparation of a preliminary mixture in Banbury internal mixers, this mixture is feed onto roll mills in order to obtain a
5 laminate of specific thickness that, when cut in a pelletiser, produces a cube-shaped granule.

The end result of all three of the above methods is that a compound in granule form is obtained, ready for transforming by the end user in extrusion or injection moulding processes.

10 STAGE TWO involves the transformation of the product from a thermoplastic into a final thermoset polymer.

To do this the compound is fed in an injection press for plastics comprising a punching screw injector consisting of a cylinder and a screw suited to feeding the granules into the cylinder which, when heated to the required temperature,
15 melts or plastifies the compound.

By appropriately controlling the cylinder's temperature so that it does not reach initiation temperature, that is usually around 110 – 130°C, of both the cross-linking agent and the foaming agent, the cycle can be performed within the material's thermoplastic range.

20 The now molten compound can then be injected into the mould which, if kept at a low temperature (i.e. 20 - 60°C), does not initiate the cross-linking and foaming processes while, if heated to a high temperature of around 150 – 190°C for sufficient time, it heats the polymer initiating peroxide activity, thereby starting up the cross-linking process in the polymer and transformation
25 of the foaming agents into gas.

Even though the gas develops a high pressure, it is confined within the mould by the clamping force of the press, thereby remaining finely dispersed throughout the polymer.

30 The cross-linking process then progressively proceeds according to the established rheological graph in the following stages:

- decomposition of the peroxide with formation of free radicals;
- extraction of hydrogen atoms by the free radicals from the polymer chains that gives rise to products of decomposition of the stable peroxides and a polymer with free radicals;
- 35 - combination of the radicals linked to the polymer and formation of cross-

linking.

As is known, both quantity and type of peroxide added will control the degree of cross-linking.

In particular, if the stoichiometric quantity of peroxide added to the mixture is
5 sufficient total cross-linking of the polymer can be achieved otherwise cross-linking levels will be less than 100%.

By selecting the right temperature and polymer curing time in the mould, the cross-linking reaction is completed only when all the peroxide is consumed thereby preventing a subsequent, further increase in viscosity, so that partial or
10 total cross-linking is achieved.

On conclusion of the cross-linking process the mould is opened allowing the gases to suddenly expand the product, making it eject automatically from the mould with a kind of small explosion.

The quantity of foaming agent in the compound, and consequently the gas
15 volume generated, together with the resilience and the resistance to the gas expansion pressure offered by the polymer determine the amount of volume increase in the product.

To be more precise the volume of the final cross-linked product is directly proportional to the gas volume generated, the physical and mechanical
20 properties of the cross-linked polymer and the level of cross-linking reached by the polymer.

The foamed product is characterised by an expanded core and a very smooth, compact outer surface due to the greater level of cross-linking and therefore resilience that the part of surface polymer in contact with the mould reaches
25 first.

The product appears with a compact surface, brilliant appearance and coloured depending on both the types and quantity of pigments in the compound.

Its shape traces the mould's form even though it is proportionally larger in size, depending on the foaming agents in the compound less shrinkage due to
30 thermal contraction created by the difference in temperature between mould and environment.

A fundamental characteristic of the cross-linked products described above is their softness which, together with their specific weight, enable extremely
35 lightweight materials to be obtained, capable of substituting more expensive

products such as cork, rubber, etc.

Known compounds have a formulation given by weight for the mixture described in the table below:

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene – Master 40% in EVA	2
Azodicarbonammide 50 % in ZnO	4.5
Zn Stearate	0.5
Stearin	1
Master carbon black 50% in EVA	2

5

The mixture blended in a Ribbon blender and extruded with a twin-screw extruder at a temperature of 100 – 110 °C, produces cylindrical granules approx. 3 mm in diameter and 2.5 mm in length which, moulded in a punching screw injection press fitted with the mould of a plate approx. 6 mm thick, 90 mm in width and 120 mm in length, heated to 180 °C with a cylinder temperature of 95°C and a cross-link curing time of 4 minutes, produce a parallelepiped cross-linked, foamed product.

Some experimental measurements made on the foamed plate are given below that, against known parameters, will allow to evaluate certain mechanical properties of the final product:

15

Density	g/cm ³	200	ASTM D 792
Hardness	Asker C 23°C	62	
Tensile strength	Mpa	2.5	ASTM D 638
Elongation at break	%	250	ASTM D 638
Gloss measurement 60° (a)	%	16	ASTM D 523

In particular it should be noted the measurements made on the product obtained from the compound described previously, using a ZEHNTER-GLOSSMETER reflectometer at a 60° light angle. The value given was 16, which is directly proportional to the amount of light reflected.

20

This enables us to highlight a great limitation of these cross-linked, foamed products arising from their intrinsic gloss that does not permit their use in certain sectors such as for instance footwear.

5 In fact, these sectors use cross-linked foamed products to make soles for shoes or slippers as an alternative to materials such as leather, rubber or cork, which are considered more valued materials and therefore more expensive. These, as is known, are intrinsically matt that allows them to be easily distinguished from cross-linked foamed products, used as an alternative since they cost less, but having a characteristic gloss.

10 Another inconvenience in these foamed products is their thermal dimensional instability, in particular when these materials are taken to high temperatures as they tend to shrink.

In fact these products can, whether accidentally or for reasons of application, be exposed to greater temperatures than environmental temperature. As a
15 consequence these materials can incur permanent and irreversible shrinkage because of their thermal dimensional instability. Therefore they cannot be used in applications where dimensional stability is an essential requirement, as mentioned earlier in their use for producing slipper or shoe soles.

In fact a sole made with cross-linked material is usually glued or stitched to the
20 upper; a poor thermal dimensional stability of the sole may lead, for instance, during transport where the footwear may undergo temperatures of 50 – 70°C, to a deformed upper and as a result deformed footwear, unacceptable in appearance and with its size not corresponding to the declared size.

Therefore it seems even clearer how important it is to have a very low thermal
25 stability, possibly below 2-2.5% while maintaining adequate softness.

The scope of this invention is to overcome the limitations illustrated above.

In fact the intention is to create a compound formulation for producing foamed and compact cross-linked polyolefins that allow to manufacture matt products.

Another scope is to produce a compound formulation that allows to obtain
30 products with stability in dimension on changes in temperature compatible with the application needs, for example to produce soles, uppers or similar. Last but not least, a scope is to produce a compound formulation that allows to obtain soft products while maintaining all the aforementioned characteristics.

Said scopes are achieved by a compound for manufacturing cross-linked
35 products obtained by injection moulding that in accordance with the main claim

consists of a mixture comprising at least:

- master polymers, such as ethylene vinyl acetates (EVA), polyethylene (PE) and their mixtures, rubber, ethylene-propylene (EPR);
- cross-linking agents, such as peroxides or similar;
- 5 - various forms of foaming agents such as azodicarbonammides or similar, said compounds

characterised in that they have in addition a butadiene-acrylonitrile copolymer, even partially cross-linked.

Said scopes and advantages will be better illustrated during the description of a preferred form of execution of the invention given as a guideline but not a
10 limitation.

In fact it has been seen how the addition of butadiene-acrylonitrile copolymers, even partially cross-linked, allows to obtain compounds producing products of matt appearance with a uniform, velvety surface.

15 Butadiene-Acrylonitrile copolymers are conventionally used as solid plastifiers in modifying polymers and in order to improve their characteristics such as for example their chemical and thermal resistance.

The copolymers can have different compositions and therefore different proportions of the two monomers, in particular the types used have an
20 Acrylonitrile content of between 30 and 37%.

These products are manufactured and traded in powder form and, to prevent packing, contain additives such as fillers (calcium carbonates or silicas).

It has been seen that if these products, in other words the copolymers, are added to known cross-linking polyolefin compounds, in quantities ranging from
25 3 to 100 by weight and preferably from 5 to 30 by weight, using conventional manufacturing technology as described earlier, matt products can be obtained with a uniform, velvety surface, while the other physical properties remain basically unchanged and valid.

EXAMPLE 1

30 According to a preferred form of execution of the invention the compound, comprising the Butadiene-Acrylonitrile copolymer, has a formulation given by weight for the mixture described in the table below.

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene - Master 40% in EVA	2.2

Azodicarbonamide 50 % in ZnO	4.6
Zn Stearate	0.5
Stearin	1
Butadiene-ACN copolymer	15
Master carbon black 50% in EVA	2.2

A partially cross-linked Butadiene – Acrylonitrile copolymer with 33% ACN was used for the test. This copolymer comes in powder form, it contains approx. 9% silica as anti-packing agent and has a specific weight of 1.02 g/cm³.

- 5 The mixture blended in a Ribbon blender and extruded with a twin-screw extruder at a temperature of 100 – 110 °C has given cylindrical granules approx. 3 mm in diameter and 2.5 mm in length which, when moulded in a punching screw injection press fitted with the mould of a plate approx. 6 mm in thickness, 90 mm in width and 120 mm in length, heated to 180°C using a
 10 95°C cylinder temperature and with a cross-link curing time of 4 minutes, have produced a parallelepiped product.

The table below gives some mechanical properties obtained by experimental measurements made on the cross-linked foamed plate produced as above.

Density	g/cm ³	205	ASTM D 792
Hardness	Asker C 23°C	60	
Tensile strength	Mpa	2.1	ASTM D 638
Elongation at break	%	260	ASTM D 638
gloss measurement 60° (a)		6	ASTM D 523

15

Thermal stability on 120x90X6 mm plate	
Shrinkage 24 h at 70°C	1.8 %
Shrinkage 24 h at 50°C	0.6 %

It is important to note how the addition of copolymer in the compound has provided a product that, when put to the gloss measurement with a 60° light angle, has a gloss of 6 or in other words nearly a third of the value obtained with the aforementioned known compounds.

EXAMPLE 2

According to a preferred form of execution of the invention the compound, comprising the copolymer, has a formulation given by weight for the mixture described in the table below.

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene – Master 40% in EVA	2.4
Azodicarbonammide 50 % in ZnO	4.7
Zn Stearate	0.5
Stearin	1
Butadiene-ACN copolymer	30
Master carbon black in EVA	2.4

This test used the same Butadiene–Acrylonitrile (ACN) copolymer with 33% ACN as in example 1 with the difference that its quantity was increased from 15 to 30 by weight.

Density	g/cm ³	205	ASTM D 792
Hardness	Asker C 23°C	58	
Tensile strength	Mpa	2.2	ASTM D 638
Elongation at break	%	250	ASTM D 638
gloss measurement 60° (a)		3	ASTM D 523

Thermal stability on 120x90X6 mm plate	
Shrinkage 24 h at 70°C	1.7 %
Shrinkage 24 h at 50°C	0.6 %

It is important to note how the increase by weight of copolymer in the compound has, against example 1, further lowered the gloss measurement. In particular it can be seen that if the copolymer content is doubled by weight then light reflection is halved.

5 EXAMPLE 3

The same composition used in example 2 has been repeated adding the styrene-ethylene-propylene-styrene (SEPS) block copolymer.

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene – Master 40% in EVA	2.5
Azodicarbonamide 50 % in ZnO	4.75
Zn Stearate	0.5
Stearin	1
SEPS	10
Butadiene - ACN copolymer	30
Master carbon black 50% in EVA	2.0

- 10 It has in fact been seen that the addition of Styrene–Ethylene–Propylene–Styrene (SEPS) type block copolymers, in quantities ranging from 1 to 50 by weight and preferably between 3 and 20 by weight, gives compounds generating products with lower hardness and greater thermal dimensional stability without altering its matt appearance.
- 15 These thermoplastic polymers (SEPS) are obtained by hydrogenation of known Styrene–Isoprene–Styrene (SIS) based thermoplastic rubbers. The thermoplasticity of these elastomers is tied to the styrenic group, or rather polystyrenic, which if heated melts and allows the polymer to be transformed by traditional thermoplastic processes.
- 20 So SEPS have a thermoplastic nature and because of the hydrogenation process they have a saturated structure and their resistance to ageing and oxidation is therefore commonly accepted. Their function is to elastomerise and soften the compound without taking part in cross-linking.
- 25 The SEPS block copolymer used has the following characteristics:

Styrene content	13 %
Hardness	38 Shore A
Tensile strength	11.8 Mpa
Elongation at break	1110 %
Physical form	Granules

The normal procedure gives the following characteristics:

Density	g/cm ³	200	ASTM D 792
Hardness	Asker C 23°C	56	
Tensile strength	Mpa	2.2	ASTM D 638
Elongation at break	%	250	ASTM D 638
Gloss measurement 60° (a)		3	ASTM D 523

5

Thermal stability on 120x90X6 mm plate	
Shrinkage 24 h at 70°C	1.8 %
Shrinkage 24 h at 50°C	0.6 %

As mentioned earlier the addition of the SEPS copolymer has allowed to obtain a reduction in product hardness that, as can be seen by a comparison of the relevant table for example 2, has gone from 58 to 56 ASKER C 23°C. Similarly thermal stability is well within the desired limits.

10

EXAMPLE 4

The same composition used in example 1 has been repeated adding the Styrene-Ethylene-Butylene-Styrene (SEBS) block copolymer.

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene – Master 40% in EVA	2.6
Azodicarbonamide 50 % in ZnO	4.75
Zn Stearate	0.5
Stearin	1
SEBS	10

Butadiene – ACN copolymer	30
Master carbon black 50% in EVA	2.6

In fact it has been seen that the addition of Styrene–Ethylene–Butylene–Styrene (SEBS) block copolymers in quantities ranging from 1 to 50 by weight and preferably between 3 and 20 by weight, allows to obtain compounds producing products with lower hardness without altering thermal dimensional stability of the initial polyolefinic polymer.

These thermoplastic elastomers (SEBS) are obtained by hydrogenation of SBS (Styrene–Butadiene– Styrene).

The thermoplasticity of these elastomers is tied to the styrenic group, or rather polystyrenic, which if heated melts and allows the polymer to be transformed by traditional thermoplastic processes.

So SEBS have a thermoplastic nature and because of the hydrogenation process they have a saturated structure and their resistance to ageing and oxidation is therefore commonly accepted.

Their function is to elastomerise and soften the compound without taking part in cross-linking.

The SEBS block copolymer used has the following characteristics:

Styrene content	12 %
Hardness	46 Shore A
Tensile strength	2.7 Mpa
Elongation at break	980 %
Physical form	Granules

The normal procedure gave the following characteristics:

Density	g/cm ³	200	ASTM D 792
Hardness	Asker C 23°C	56	
Tensile strength	Mpa	2.1	ASTM D 638
Elongation at break	%	255	ASTM D 638
Gloss measurement 60° (a)		3	ASTM D 523

Thermal stability on 120x90X6 mm plate	
Shrinkage 24 h at 70°C	1.7 %
Shrinkage 24 h at 50°C	0.5 %

As mentioned earlier the addition of the SEBS copolymer has allowed to obtain a reduction in product hardness that, as can be seen by a comparison of the relevant table for example 2, has gone from 58 to 56 ASKER C 23°C. Similarly
5 thermal stability is well within the desired limits.

CLAIMS

1) Compound for manufacturing cross-linked products with great softness and thermal stability made by injection moulding, consisting of a mixture comprising at least:

- 5 - master polymers such as ethylene vinyl acetates (EVA), or polyethylene (PE) and their mixtures, otherwise ethylene-propylene rubber (EPR);
- cross-linking agents, such as peroxides or similar;
- foaming agents, such as azodicarbonammides or similar

10 **characterised in that** it has in addition butadiene-acrylonitrile copolymers, even partially cross-linked.

2) Compound according to claim 1) **characterised in that** it contains said copolymers in quantities by weight ranging from 3 to 100 and preferably between 5 and 30 by weight.

15 3) Compound according to claim 1) **characterised in that** it has the following formulation:

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene	2.2
– Master 40% in EVA	
Azodicarbonammide 50 % in ZnO	4.6
Zn Stearate	0.5
Stearin	1
Butadiene – ACN copolymer	15
Master carbon black 50% in EVA	2.2

4) Compound according to claim 1) **characterised in that** it has the following formulation:

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene	2.4
– Master 40% in EVA	
Azodicarbonammide al 50 % in ZnO	4.7
Zn Stearate	0.5
Stearin	1
Butadiene - ACN copolymer	30
Master carbon black 50% in EVA	2.4

5) Compound according to claim 1) **characterised in that** it comprises Styrene-Ethylene-Propylene-Styrene (SEPS) type block copolymers in quantities ranging from 1 to 50 by weight and preferably between 3 and 20 by weight

5 6) Compound according to claim 1) **characterised in that** it comprises Styrene-Butadiene-Styrene (SEBS) type block copolymers in quantities ranging from 1 to 50 by weight and preferably between 3 and 20 by weight

7) Compound according to claim 1) **characterised in that** it comprises Styrene-Ethylene-Propylene-Styrene (SEPS) type block copolymers and
10 Styrene- Butadiene- Styrene (SEBS) type block copolymers in quantities ranging from 1 to 50 by weight and preferably between 3 and 20 by weight.

8) Compound according to claim 5) **characterised in that** it has the following formulation:

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene	2.5
– Master 40% in EVA	
Azodicarbonammide 50 % in ZnO	4.75
Zn Stearate	0.5
Stearin	1
SEPS	10
Butadiene – ACN copolymer	30
Master carbon black in EVA	2.6

15 9) Compound according to claim 6) **characterised in that** it has the following formulation:

EVA 2MFI 18 % VA	100
Silica amorphous powder	5
Bis (tert-butylperoxypropyl) benzene	2.5
– Master 40% in EVA	
Azodicarbonammide 50 % in ZnO	4.75
Zn Stearate	0.5
Stearin	1

SEBS	10
Butadiene – ACN copolymer	30
Master carbon black in EVA	2.6

10) Compound according to any one of the above claims **characterised in that** said copolymer has the following Butadiene–Acrylonitrile (ACN) copolymer with 33% ACN, in powder, approx. 9% of silica as anti-packing agent, specific weight 1.02 g/cm³.

11) Compound according to claim 5) or 7) or 8) **characterised in that** said SEPS type block copolymer has the following properties:

Styrene content	13 %
Hardness	38 Shore A
Tensile strength	11.8 Mpa
Elongation at break	1110 %
Physical form	Granules

12) Compound according to claim 6) or 7) or 9) **characterised in that it** has the following properties:

Styrene content	12 %
Hardness	46 Shore A
Tensile strength	2.7 Mpa
Elongation at break	980 %
Physical form	Granules

INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 00/08668

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08L23/08

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 812 225 A (K.HOSODA ET AL) 21 May 1974 (1974-05-21) example 48	1, 2, 11, 12
A	DATABASE WPI Derwent Publications Ltd., London, GB; AN 1995-136958 XP002167263 & JP 07 062130 A (TOYO KUORITIWAN), 7 March 1995 (1995-03-07) abstract; example 3	1-12
A	US 5 635 562 A (MALCOLM COLLIN C) 3 June 1997 (1997-06-03) table II	1-12

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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